Phonons and High-Temperature Superconductivity

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Introduction

Superconductivity—the loss of all electrical resistance in a substance—mystified scientists after its early discovery in 1911 when Dutch physicist Heike Kammerlingh Onnes observed that mercury exhibited unique properties at very low temperatures. By removing heat and thus reducing the normal vibration of the atoms in the metal, Onnes expected a *steady* decrease in electrical resistance. What he found instead was the disappearance of electrical resistance altogether at a characteristic transition temperature (T_c) at which the metal became superconducting! As temperatures were pushed lower and lower, many more metals and alloys were added to a growing list of materials thought to have potentially widespread applications. However, applying this phenomenon to practical solutions requires the design of materials that can attain superconductivity at or near room temperature. With this goal in mind, researchers at LANSCE are studying a new class of high-temperature superconductors (HTSCs),[1] which attain electrical resistance beyond 100 K (e.g., 273 K is the melting point of ice, and 293 K is room temperature). Unlike *low-temperature* superconductors, HTSCs are not well understood, particularly in terms of the Bardeen-Cooper-Schrieffer (BCS) microscopic theory of superconductivity, which quantum mechanically describes how conduction electrons pair together to reduce the total energy of a system. LANSCE researchers are seeking answers to fundamental physics questions about HTSCs, in particular, how electrons move about in a strongly correlated electron system. These answers will inevitably lead to a fundamental understanding of how HTSCs work and perhaps eventually to the design of superconductors with T_cs closer to room temperature than those of their forerunners.

Understanding High-Temperature Superconductors

Understanding HTSC materials is a challenging problem because they are not truly metals. HTSC materials epitomize the general class of strongly correlated electron materials where the mutual repulsion of electrons dominates the physics. HTSC materials are all based on insulating and magnetic copper-oxide *parent* compounds like La_2CuO_4. The electrical conductivity of this material is controlled by adding impurities such as strontium to the parent compound (e.g., $La_{1.85}Sr_{0.15}CuO_4$)—a procedure reminiscent of the semiconductor process known as *doping*. Doping introduces "charge carriers" called *holes* into the system. Holes signify the absence of electrons in the system and are responsible for the electrical conductivity of the material. But unlike semiconductors, the doped holes in HTSCs migrate to two-dimensional CuO_2 sheets, a structural element common to all cuprate (copper oxide) superconductors, and are strongly correlated to each other and also coupled to magnetic copper spins and lattice displacements. And so the position and motion of a hole depends on the position of other holes, copper spin orientations, and atomic positions! Well-understood normal metals establish the framework for implementing the BCS theory, but the intermingling of charge, spin, and lattice degrees of freedom in HTSC systems typifies the complexity of these new materials.

One approach is to imagine that the doped holes phase segregate into hole-rich (metallic) and hole-poor (insulating and magnetic) regions.[2] At low-doping concentrations where hole-rich regions are well separated, the material remains an insulator because significant energy is expended as the hole creates spin disturbances and lattice deformations while traveling through a hole-poor region. As the number of holes in a system increases, the phase-segregated regions become interconnected and are thought to organize themselves loosely into parallel strands called "stripes" that allow for hole conductivity throughout the material. The best experimental evidence for stripes comes from the observation of quasi-static spin structure by inelastic neutron scattering and its interpretation as a slowly moving, regular arrangement of hole-poor regions.[3] This implies a slowly moving, regular arrangement of chains of holes separated by 4a (where a is the in-plane lattice constant). However, these measurements can also be interpreted with an itinerant (or homogeneous) model of the holes. In materials such as insulating nickelates and non-superconducting cuprate compounds, stripes are static, and both hole and spin arrangements are observed simultaneously by neutron and x-ray diffraction.[4] In the cuprate superconductors, where holes are moving around, the hole arrangement within the stripe picture has not been observed.

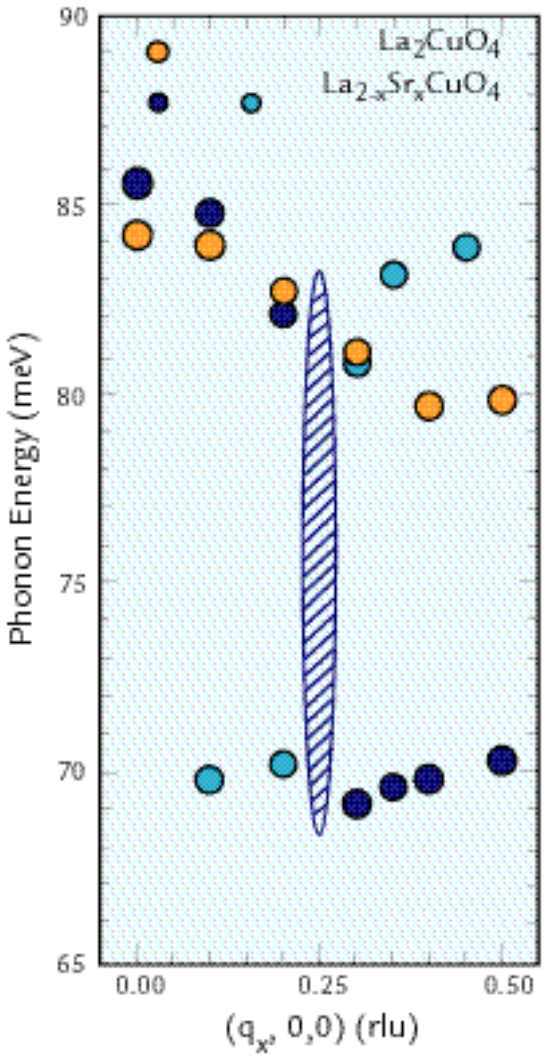
Our research collaboration is trying to determine the hole arrangements in HTSCs indirectly by measuring the dispersion of phonons. Phonons are quanta of the vibrations in a crystal lattice that (in the appropriate frequency range) describe classical sound.

Phonon "modes" can be measured with inelastic neutron scattering—a technique that simultaneously measures momentum and energy transfer to a material and therefore maps out the phonon dispersion $\omega(\mathbf{q})$ (the relationship between the direction and wavelength, or wave vector, of a lattice displacement field and its frequency). Because the lattice vibrations are coupled to the holes, it is possible that hole arrangements are reflected in the details of the phonon dispersion. Anomalies in the phonon dispersion at a particular wave vector can correspond to spatial hole correlations with the same wave vector.

The phonon anomalies in the cuprates present themselves as reductions in the frequency and lifetime of particular oxygen modes with hole doping. An example of phonon softening is shown in Fig. 1 for the La_2CuO_4 system. The frequency softening with doping (12-20%) is extremely large and points to a huge electron-lattice coupling. The current body of measurements already attests to the ubiquitous nature of the oxygen phonon anomalies in HTSCs. They have been observed in several superconducting compounds with different crystal structures and methods of introducing holes. Our research is aimed at understanding the systematic effects of hole concentration and temperature on the lattice dynamics of several HTSC compounds. Our recent experimental results are suggestive of phase segregation and local charge fluctuations that may be consistent with the stripe model.

Using Inelastic Neutron Scattering to Measure the Phonon Spectrum

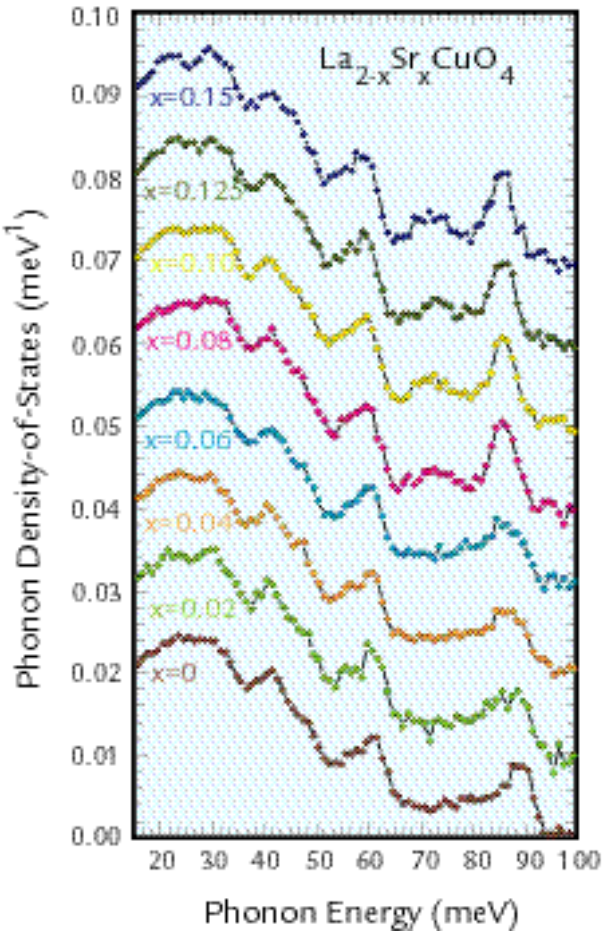
Using triple-axis spectrometers at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory, we mapped out the dispersion of phonons $\hbar\omega(\mathbf{q})$ along particular directions in single-crystal samples using inelastic neutron scattering. We also studied powder samples with the time-of-flight technique using LRMECS (Low-Resolution Medium Energy Chopper Spectrometer) at Argonne National Laboratory's Intense Pulsed Neutron Source. Powder data, which average $\hbar\omega(\mathbf{q})$ over all directions of q, are collected and summed over a wide range of scattering angles, which averages over all magnitudes $|\mathbf{q}|$, to give the phonon density-of-states $Z(\omega)$. The triple-axis data on single crystals give information about specific phonon modes and branches and is used to study the detailed behavior of the lattice dynamics of HTSC materials. The powder time-of-flight data give a rough overview of the lattice dynamics; however, it is more amenable to parametric studies of composition dependence.



▲ ***Fig. 1.*** *The phonon dispersion of a high-frequency oxygen-phonon branch at T=10 K in undoped and doped $La_{2-x}Sr_xCuO_4$. By doping, large softening occurs between* $\mathbf{q}$ *= (0.25,0,0) and (0.5,0,0), and the phonon mode at (0.25,0,0) is strongly broadened. Additionally, weak "extra" modes are observed by doping.*

Results

Fig. 2 shows the phonon density-of-states for several different hole concentrations of $La_{2-x}Sr_xCuO_4$. As more holes are added (higher x), a band of phonons develops near 70 meV. This band of phonons contains the same phonons whose frequency softened from 80 meV, as shown in Fig. 1. The 70-meV soft band occurs only above hole concentrations $x > 0.06$. The hole concentration $x = 0.06$ is near the critical concentration where the hole-doped insulator begins to show electrical conductivity (the metal-insulator transition). Thus, the mobility of the holes affects the lattice dynamics. For $x > 0.06$, the soft band that appears occurs only at

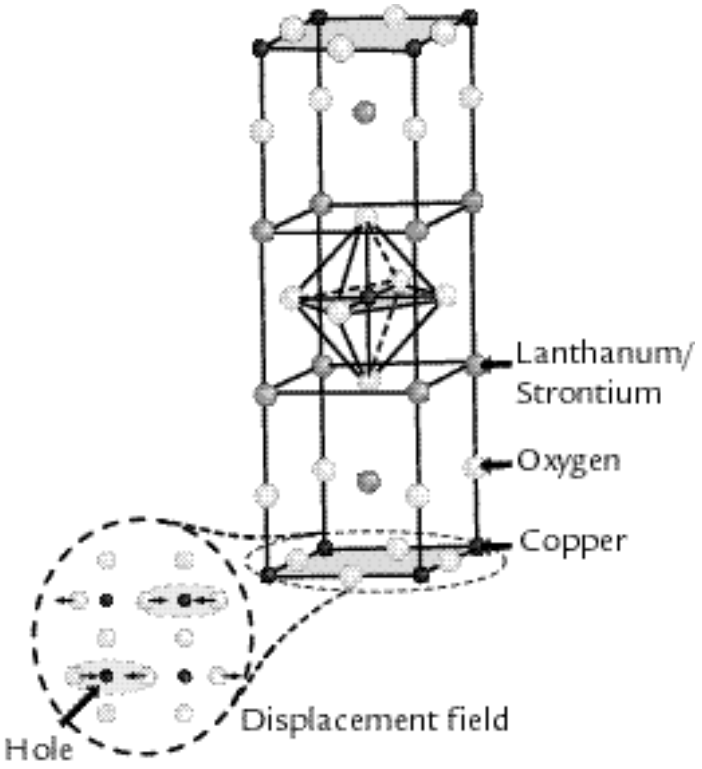


▲ ***Fig. 2.*** *The phonon density-of-states at T = 10 K for $La_{2-x}Sr_xCuO_4$ as a function of hole doping. Between 0.06 < x < 0.08 and close to the metal-insulator transition in this material, a soft phonon band suddenly occurs at 70 meV. This softening is also observed in single-crystal phonon dispersion measurements (see Fig.1).*

70 meV. There is no gradual softening of the anomalous phonon modes with doping, as one might expect from a gradual change in the lattice parameters or the interatomic potential (as holes screen the ions). Rather, the oxygen modes behave as if they exist in two distinct environments. The intensity of the developed soft band is quite large, which implies that a large number of phonons (~ 5% to 10% of all possible oxygen modes) are affected. As we will see from the single-crystal data (and also evident in Fig. 1), the large number of oxygen modes in the soft band arise from a specific pocket in **q**-space. This "wave packet" of phonons can equally be interpreted as a lattice vibration localized on the lattice, similar to the vibrations of a molecule in a loosely bound molecular crystal. The phonon anomalies thus result from electron-lattice coupling from local charge fluctuations in the hole-rich regions.

Using single crystals, we can probe more deeply into the oxygen-phonon anomalies.[5] Fig. 1 shows that the phonon branch along the (100)-direction in $La_{1.85}Sr_{0.15}CuO_4$ (Cu-O-Cu direction in the CuO_2 plane) is nearly discontinuous halfway to the zone boundary. The softening occurs in a pocket in **q**-space centered at **q**a/2p=(0.5,0,0) and extends to (0.25,0,0) and (0.5,0.15,0) in the plane. In this region, the dispersion is flat, again signifying localized vibrations.

The discontinuity at $\mathbf{q}_x a/2\pi = 0.25$ in these compounds gives additional information about the interaction between the holes and phonons. The wave vector of the discontinuity is consistent with the hole spacing of 4a as measured by the dynamic spin structure. However, simple modeling of the lattice dynamics supports a picture where the phonons are coupled to holes that are fluctuating back and forth on a length scale of 2a. This discrepancy in the interpretation of the phonon anomaly has yet to be resolved, but in either case an inhomogeneous distribution of holes is required, as shown in Fig. 3. Recent results show the discontinuity beginning to disappear at a characteristic temperature that depends on the hole concentration. At this temperature, holes may begin to delocalize throughout the



▲ ***Fig. 3.*** *The structure of La_2CuO_4. Holes created when La is replaced by Sr migrate into the CuO_2 plane (shaded in gray). Phonon measurements suggest that the holes are also localized and interact with the phonons, as shown in the inset.*

system, destroying the special electron-lattice coupling that depends on local charge fluctuations. This characteristic temperature, T*, has also been observed through other techniques.

We are beginning to understand how electron-lattice coupling causes phonon anomalies in the cuprate materials. The phonons are best understood as localized phonons in a two-phase model, implying that the holes are microscopically phase segregated and *not* itinerant. Strong electron-lattice coupling only occurs when the doped holes are mobile. The phonon measurements are therefore consistent with some aspects of the stripe model, such as phase separation; however appropriate length scale (2a or 4a) seems to be at odds.

References

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